REPORT DOCUMENTATION PAGE

Form Approved OMB No. 0704-0188

1. REPORT DATE *(DD-MM-YYYY)*
26-04-2005

2. REPORT TYPE
Final Report

3. DATES COVERED *(From – To)*
15 March 2004 - 15-Mar-05

4. TITLE AND SUBTITLE
Mechanisms Of Iodine Dissociation In Chemical Oxygen Iodine Lasers

5a. CONTRACT NUMBER
FA8655-04-1-3031

5b. GRANT NUMBER

5c. PROGRAM ELEMENT NUMBER

6. AUTHOR(S)
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5d. PROJECT NUMBER

5d. TASK NUMBER

5e. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
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8. PERFORMING ORGANIZATION REPORT NUMBER
N/A

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
EOARD
PSC 802 BOX 14
FPO 09499-0014

10. SPONSOR/MONITOR'S ACRONYM(S)

11. SPONSOR/MONITOR'S REPORT NUMBER(S)
SPC 04-3031

12. DISTRIBUTION/AVAILABILITY STATEMENT
Approved for public release; distribution is unlimited.

13. SUPPLEMENTARY NOTES

14. ABSTRACT

This report results from a contract tasking Ben-Gurion University of the Negev as follows: The Grantee will investigate the dissociation mechanism in the COIL using modern experimental methods based on sensitive absorption spectroscopy by tunable diode lasers. The main issues to be addressed are the following:
1. Determination of the kinetic mechanism of I2 dissociation in the COIL and the rate constants of key elementary reactions.
2. Experimental measurements and modeling of the fundamental I2 dissociation parameter N equal to the average number of O2(1D) molecules needed to dissociate one molecule of I2.
3. Assessment the role of the metastable electronically excited states of I2 (the A3P1 and A'3P2 states) for I2 dissociation using pulsed laser pump-probe techniques

15. SUBJECT TERMS
EOARD, Chemical oxygen iodine lasers

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18, NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			DONALD J SMITH
UNCLAS	UNCLAS	UNCLAS	UL	13	19b. TELEPHONE NUMBER *(Include area code)* +44 (0)20 7514 4953

Standard Form 298 (Rev. 8/98)
Prescribed by ANSI Std. Z39-18

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Mechanisms of iodine dissociation in chemical oxygen iodine lasers

Contract No. FA8655-04-1-3031

BGU Internal Contract No. 82242101

Final Report for the Period

15.03.2004 - 14.03.2005

Principal Investigators: Dr. Boris Barmashenko and Prof. Zamik Rosenwaks

Content

I. Abstract

Due to the success in raising the chemical efficiency, η_{chem}, of our supersonic COIL to a new record (~ 40%), approaching the theoretical limit for this efficiency, we have devoted most of our efforts during the reported period to studying the optimal conditions for lasing and detailed diagnostic study of the $O_2(^1\Delta)$ yield and spatial distributions of the gain and temperature in the resonator. These diagnostic studies are a prerequisite for the measurements of the I_2 dissociation fraction which are underway.

The following projects have been carried out during the reported period:

1. Measurements of the lasing power for new supersonic nozzles with different injection location along the flow and for different throat heights and achievement of 40% chemical efficiency.
2. Measurements of the gain and temperature for different nozzles with supersonic mixing.
3. Measurements of the $O_2(^1\Delta)$ yield and chlorine utilization in the singlet oxygen generator.
4. Design, manufacturing and testing of a new optical system for measurements of $[I_2]$ using 488 nm probe beam from a blue laser.

II. Description of the results

1. Measurements of the lasing power for new supersonic nozzles with different injection location along the flow and different throat heights and achievement the record-breaking 40% chemical efficiency

To optimize the supersonic mixing scheme we measured the power for a series of new supersonic nozzles with different locations of the iodine injection holes. Some of these nozzles were profiled to avoid shock waves and they have different locations of the iodine injection holes in the supersonic section of the flow. It is also possible to change the nozzle throat height (5 mm or 8 mm). For these nozzles a series of experiments was carried out where we measured lasing power in order to find the nozzle for which the power is maximal (see Table I). We used nozzle No.3 for which we got 33% chemical efficiency[6] as a reference. In our experiments we tested four new nozzles. Three of the nozzles were profiled with a 45^0 angle between the primary and secondary flow and different locations of non-staggered injection holes along the flow. The flow height H at the injection location is 6.5, 7.5 and 8.75 mm for nozzles No. 4, 5 and 6, respectively. A non-profiled nozzle, No. 7, with a throat height of 8 mm, larger than the 5 mm throat height of nozzles No. 1-6, was also tested.

To achieve additional increase of η_{chem} we decreased the temperature of the secondary flow by stopping the electrical heating of the iodine-oxygen mixing system which was applied in our previous experiments. As a result, the highest gain and power were obtained for smaller iodine flow rate, nI_2, as compared to the nI_2 needed to get the highest gain and power when heating was applied. Due to the higher nI_2 needed for maximum gain and power in the latter case, their values were lower than in the former case for the same flow rates of the other reagents.

Table I. Maximum power and chemical efficiency for different supersonic mixing schemes. Nozzles No. 3 – 6 have 5 mm throat height; nozzle No.3 has staggered injection holes, and nozzles No. 4-6 have non-staggered injection holes

Run No.	Nozzle number and description	nCl_2 [mmole/s]	nN_2 [mmole/s]	nI_2 [mmole/s]	Pressure in the JSOG [Torr]	Pressure in the resonator [Torr]	Power [W]	Chemical efficiency [%]
1	No.3, reference	17.4	28	0.6	14.6	2.4	517	32.7
2	No.4, profiled, H = 6.25 mm	17.3	17	0.31	14.3	2.0	587	37.2
3	No.5, profiled, H = 7.5 mm	17.4	28	0.39	15.5	2.3	600	37.9
4	No.6, profiled, H = 8.75 mm	17.3	28	0.36	14.6	2.2	553	35
5	**No.7, non-profiled, 8 mm throat height**	**17.4**	**28**	**0.45**	**13.3**	**2.3**	**627**	**39.6**

The results of the power measurements are presented in Table I. The maximum values of the chemical efficiencies obtained for profiled nozzles Nos. 4, 5 and 6 with 5 mm throat height (runs 2 to 4, respectively) are ~ 37%, 38% and 35%, respectively. They are larger than the ~ 33% efficiency achieved for nozzle 3 in (run 1, V. Rybalkin, A. Katz, B. D. Barmashenko, and S. Rosenwaks, *Appl. Phys. Lett* 82, 3838 (2003)). It was difficult to find which of the nozzles (No. 4 – 6) is best since the power strongly depends on both the mirror quality which tends to deteriorate from run to run and on the mirrors' alignment. Analysis of other results (not presented in Table I) shows that for the nozzles with 5 mm throat height, the dependence of the power and chemical efficiency on the injection location along the flow is very weak. However, the power obtained in run 5 for nozzle No.7 with 8 mm throat height was higher than that obtained in run 3 for nozzle No. 5 with a smaller (5 mm) throat height. In both runs we used the same mirrors with total transmission T = 1%. Hence, increase of the nozzle throat height results in power increase. The reason for higher power obtained for nozzle No.7 is smaller generator pressure due to larger throat height resulting in smaller energy losses of singlet oxygen.

Maximum power of 627 W with chemical efficiency of about 40% was achieved for nozzle No. 7. 40% efficiency is the highest reported chemical efficiency of a supersonic COIL. It is much larger than the 33% efficiency reported recently by us in [1] and is close to the upper theoretical limit of the COIL chemical efficiency.

The nozzle throat height strongly affects the power and η_{chem}. We found that η_{chem} increased from ~ 38 to ~ 40% as the throat height increased from 5 to 8 mm. To find if further increase of the throat height results in the efficiency increase we manufactured a nozzle with 10 mm throat height, i.e., without geometrical throat. The throat in such a nozzle is created by the jets injected at 45^0 to the primary flow. Power measurements, however, showed that η_{chem} for this nozzle is only about 31%, i.e., is smaller than for both 5 and 8 mm throat height. That means that the optimal throat height is about 8 mm.

The temporal evolution of the power and the chemical efficiency, as well as of $n\mathrm{Cl}_2$, is shown in Fig. 1 for conditions similar to that of the record-breaking run 5. Both the power and chemical efficiency reach maximum values and then decrease to smaller steady state values. The maximum chemical efficiency, ~ 40.0%, was measured at the early stage of operation, lasted ~ 1 s and then gradually dropped to a sustained efficiency of ~ 35.5% for ~ 20 s prior to shutting off the gas flows.

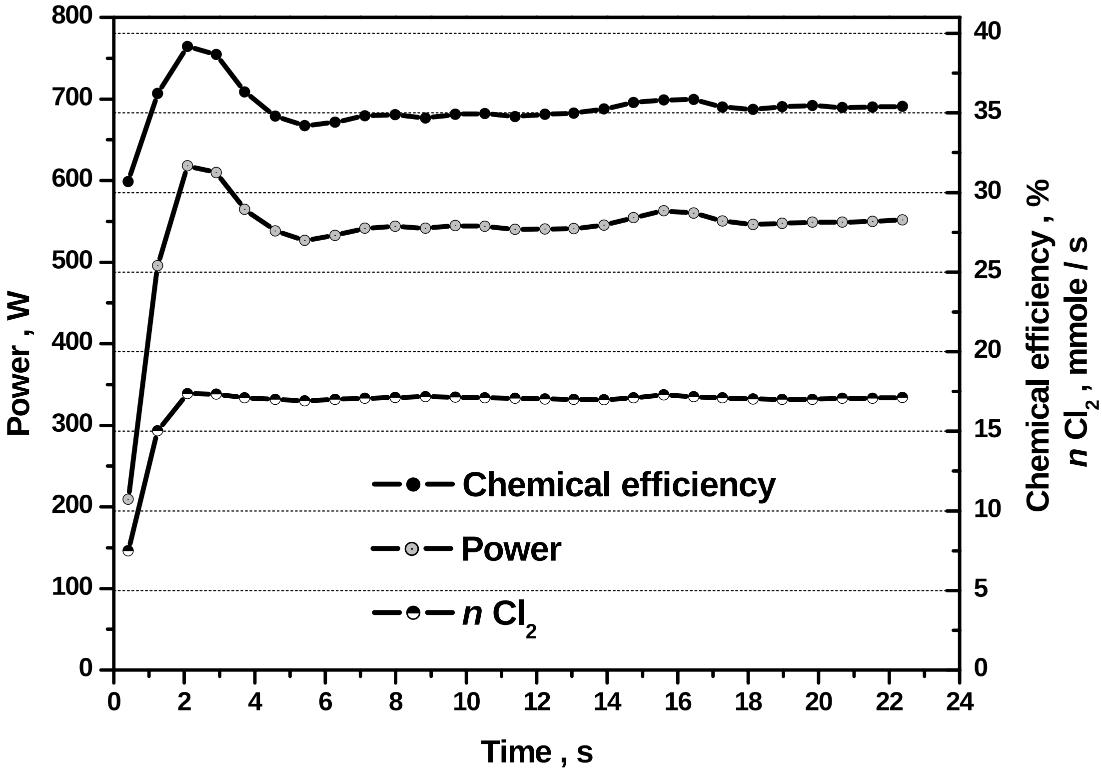

Fig. 1. Temporal behavior of the power, nCl_2 and chemical efficiency for nozzle 2c (nN_2 = 28 mmole/s, nI_2 = 0.47 mmole/s). The errors in the power, nCl_2 and chemical efficiency are ± 2 W, ± 0.1 mmole/s and ± 0.3%, respectively.

2. <u>Measurements of the gain and temperature for different nozzles with supersonic mixing</u>

Spatial distributions of the gain and temperature across the flow were measured for a non-profiled nozzle with a throat-height of 8 mm for which record-breaking efficiency (~ 40%) was achieved. Maximum values of the gain (0.7 %/cm) and minimum value of the temperature (230 K) were achieved at the flow centerline.

To achieve the highest efficiency we decreased the temperature of the secondary flow by stopping the electrical heating of the iodine-oxygen mixing system which was applied before. To find the reasons for the efficiency and power increase we measured the gain in both the heated and non-heated systems, using diode laser based diagnostic methods. Fig. 2 shows the dependence of the gain (averaged across the flow) on nI_2. It is seen that the gain measured for nI_2 corresponding to the maximum power in the non-heated system was ~ 30% higher than the gain measured for nI_2 corresponding to the maximum power in the heated system for the same flow rates of the other reagents. It is worth noting that in supersonic COILs maximum power is generally achieved at smaller values of nI_2 than for maximum gain. The values of the temperature of the gas mixture in the laser cavity for conditions of maximum power, found from the Doppler line width of the I atoms, were the same in both the heated and non-heated systems.

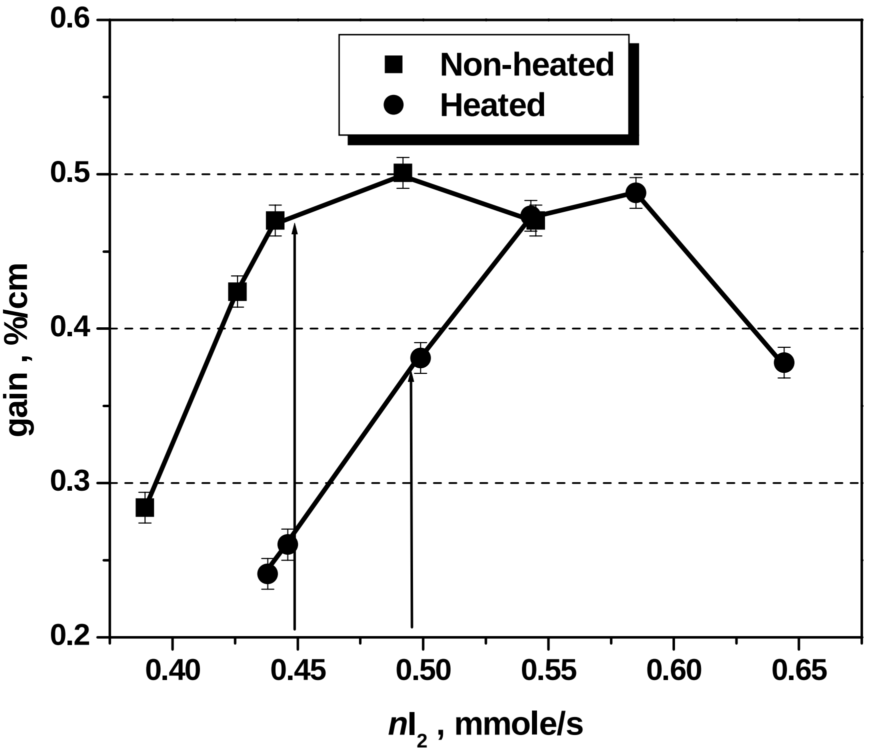

Fig. 2. Dependence of the gain on nI_2 (for the same flow rates of the other reagents) for the heated and non-heated nozzle. The arrows show the values of the gain at nI_2 corresponding to the maximum power.

The reason for the larger values of nI_2 needed for maximum power and gain in the heated system is that the heating of the secondary flow results in a smaller density of iodine species and larger flow velocity in the resonator, which, in turn, results in slower dissociation rate. To increase this rate and the iodine dissociation fraction at the resonator optical axis it is necessary to increase nI_2. Since increasing nI_2 results in increase of quenching of excited species, maximum gain and power in the heated system are lower than in the non-heated system for the same flow rates of the other reagents.

The spatial distributions of the gain and temperature across the flow were also measured for the profiled and non-profiled nozzles with 5 mm throat height and different flow heights H at the injection locations along the flow. Fig. 3 shows the dependence of the gain (averaged across the flow) on the iodine flow rate, nI_2, for these nozzles for constant chlorine flow rate ~17 mmole/s and secondary N_2 flow rates corresponding to the maximum powers achieved for these nozzles. The respective N_2 flow rates were 17 mmole/s for H = 6.25 mm and 28 mmole/s for H= 7.5 and 8.75 mm.

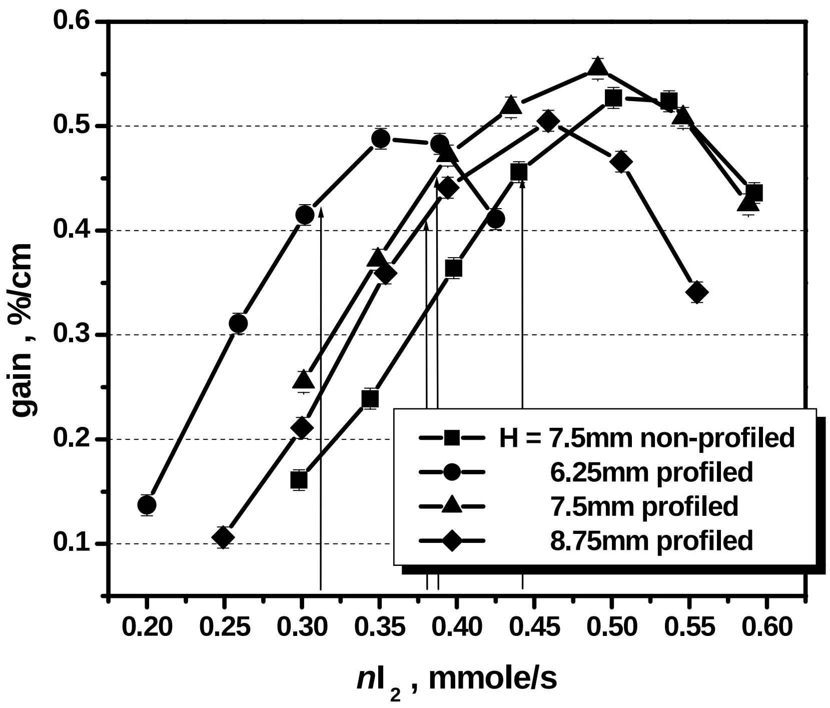

Fig. 3. Dependence of the gain on nI_2 (for the same flow rates of the other reagents) for profiled and non-profiled nozzles with a throat height h = 5 mm and different flow heights H at the injection location. The arrows show the values of the gain at nI_2 corresponding to the maximum power.

It is seen that the value of the gain is weakly dependent on H and hence on the injection location along the flow. The gain for H = 8.75 mm is a little smaller than for H =

6.25 and 7.5 mm which is in agreement with the fact that the value of the chemical efficiency η_{chem} for H = 8.75 is also smaller than for H = 6.25 and 7.5 mm. The gain and η_{chem} for non-profiled and profiled nozzles with H = 7.5 are almost the same.

The spatial distributions of the gain and temperature were measured for nozzles with different throat heights h. The dependence of the gain (averaged across the flow) on the iodine flow rate, nI_2, for these nozzles is shown in Fig. 4. It is seem that maximum gain is achieved for h = 8 mm which is in agreement with the fact that the maximum value of η_{chem} is achieved for the same throat height.

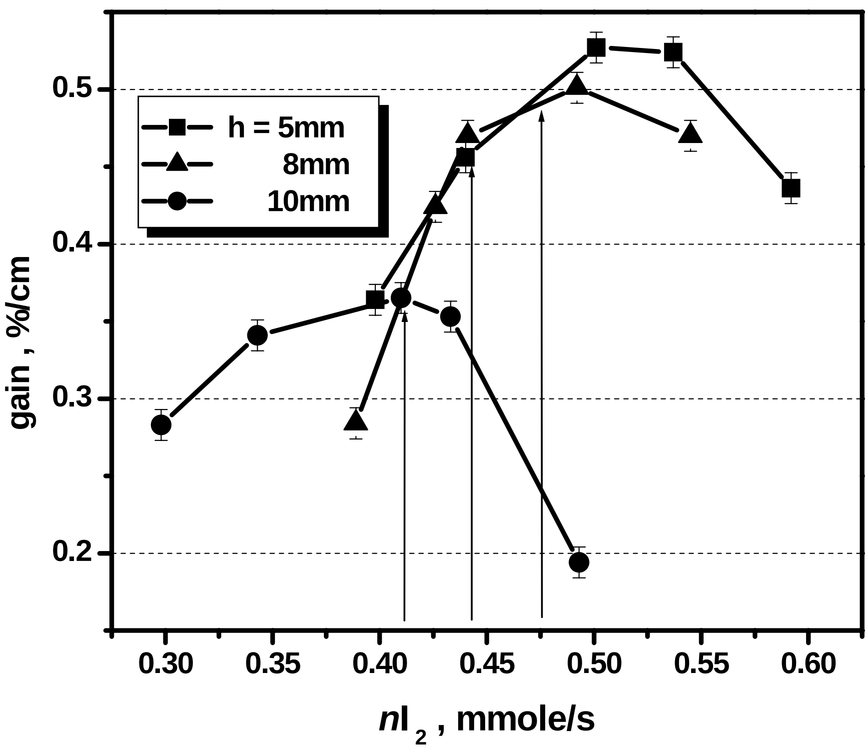

Fig. 4. Dependence of the gain on nI_2 (for the same flow rates of the other reagents) for non-profiled nozzles with different throat heights h = 5, 8 and 10 mm. The arrows show the values of the gain at nI_2 corresponding to the maximum power.

3. Measurements of the $O_2(^1\Delta)$ yield and chlorine utilization in the singlet oxygen generator

Measurements of $O_2(^1\Delta)$ yield Y and chlorine utilization U using diode laser based diagnostic system have been started. The yield depends on the energy loss of $O_2(^1\Delta)$ due to energy pooling between $O_2(^1\Delta)$ molecules in the chemical generator and is determined by the product $P_{O_2}\tau$, where P_{O_2} is the partial pressure of O_2 and τ the residence time of the flow in the generator. The smaller is $P_{O_2}\tau$ the larger is Y. To change $P_{O_2}\tau$ we changed the secondary N_2 flow rate which chokes the primary flow thus changing the generator pressure. Two nozzles with throat height h = 5 and 8 mm were used. The yield for 8 mm throat nozzle for conditions corresponding to maximum 40% efficiency is 0.73, whereas for 5 mm throat nozzle where $P_{O_2}\tau$ is larger than for 8 mm nozzle the yield is ~ 0.64. The increase of Y explains why for the 8 mm throat nozzle we got higher chemical efficiency than for the 5-mm throat nozzle. The value of the chlorine utilization is ~0.95 for both throat heights.

4. Design, manufacturing and testing of a new optical system for measurements of [I_2] using 488 nm probe beam from a blue laser.

We decided to probe [I_2] with a 488 nm beam rather than using the 540 nm green He-Ne laser beam as we did before. The reason is that the 488 nm transition of I_2 lies in the continuous part of the absorption spectrum whereas the 540 nm signal probes individual rovibronic lines of I_2. As a result the absorption cross section around 488 nm is not sensitive to variations in the temperature and ground-state rovibrational population. First we tried a blue LED as a light source, however, the signal was weaker than that of the green He-Ne and the noise was too high to perform accurate measurements of iodine in the resonator. We therefore purchased a new 488 nm solid-state laser with the same intensity as of the green He-Ne; the first tests showed the noise is very low for the new laser. A new optical system is designed and manufactured to probe the I_2 across the flow. For this system all the optical components (the collimators, beamsplitters, chopper and detectors) are moved by a linear stage across the gas flow. As a result a very accurate alignment is possible. The symmetric optical scheme is used with equal optical paths of the signal and reference beams which also resulted in a low noises. Experiments on the measurements of I_2 dissociation in the COIL are underway.

III. Summary and recommendations

A detailed diagnostic study of the ~ 40% efficient supersonic COIL was carried out. The power, small signal gain and temperature in the laser cavity were measured for different supersonic slit nozzles with both staggered and non-staggered iodine injection holes, different injection locations along the flow and different nozzle throat-heights and for heated and non-heated nozzle walls. For a given chemical generator the most important parameters affecting both the power and gain are the temperature of the nozzle walls and the nozzle throat-height. To achieve maximum power and gain the nozzle walls should not be heated. For our system there is an optimal nozzle throat-height h (~ 8 mm) corresponding to maximum power and chemical efficiency, the optimal ratio of the flow cross section areas at the nozzle exit plane and at the throat being ~ 1.25. For smaller h the generator pressure and gas residence time are too large, resulting in smaller $O_2(^1\Delta)$ yield and higher stagnation temperature, whereas for larger h the O_2/I_2 mixing efficiency is low resulting in strong decrease of η_{chem}. Other nozzle parameters, such as injection location along the flow and geometrical configuration of the injection holes, weakly affect both power and gain.

We have shown that the effects of the partial pressure of O_2 and residence time of the flow in the generator are crucial in attaining high efficiency COILs. Extremely small value of $P_{O_2}\tau$ < 0.04 Torr s achieved in our JSOG resulted in a very high $O_2(^1\Delta)$ yield, ~ 0.73. The temporal behavior of the power and η_{chem} was studied in detail. The record-breaking 40% efficiency was measured for ~ 1 s at the early stage of operation, followed by a sustained ~ 35.5% chemical efficiency for ~ 20 s. ~ 40% (and even 35.5%) is the highest reported chemical efficiency of any supersonic COIL and the closest to the theoretical limit.

Design and manufacturing of a new optical system for measurements of $[I_2]$ using 488 nm probe beam from a blue laser is finished. First experiments of the iodine dissociation are underway.

IV. Supplements

1. List of the people participating in the project

The list includes only the people participating in the research (it does not include the workers of different workshops that helped in building and maintenance of the experimental setup):

Prof. Zamik Rosenwaks, principal investigator

Dr. Boris Barmashenko, principal investigator

Mr. Victor Rybalkin, investigator (Ph. D. student)

Mr. Arje Katz, investigator (Ph. D. student)

2. List of the technical documents during the reported period

The technical documents that appeared during the reported period include one report submitted to the EOARD 6 months after the onset of the research and the papers published or submitted for publication during the reported period. Below is the list of the papers:

1. B. D. Barmashenko, V. Rybalkin, A. Katz and S. Rosenwaks, ”Parametric study of the Ben-Gurion University efficient chemical oxygen-iodine laser”, in High-Power Laser Ablation V symposium, Proc. SPIE, 2004, vol. 5448, pp.282-293.

2. S. Rosenwaks, V. Rybalkin, A. Katz and B. D. Barmashenko, ” Recent studies of Ben-Gurion University high efficiency supersonic chemical oxygen-iodine laser”, in 15th International Symposium on Gas Flow and Chemical Lasers and High Power Laser Conference, Proc. SPIE, 2004, vol. 5777 (in press).

3. V. Rybalkin, A. Katz, B. D. Barmashenko, and S. Rosenwaks, “Nearly attaining the theoretical efficiency of supersonic chemical oxygen-iodine lasers,” *Appl. Phys. Lett*., vol. 85, pp. 5851-5853, 2004.

4. V. Rybalkin, A. Katz, B. D. Barmashenko, and S. Rosenwaks, “Parametric study of a highly efficient chemical oxygen-iodine laser with supersonic mixing of iodine and oxygen ,” *IEEE JQE,* submitted for publication.

V. Acknowledgment

This material is based upon work supported by the EOARD under Contract No. FA8655-03-1-3055.